

# Nonlinear adiabatic response of interacting quantum dots

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**Abstract** –We develop a generic method in Liouville space to describe the dissipative dynamics of coherent interacting quantum dots with adiabatic time dependence beyond linear response. We show how the adiabatic response can be related to effective quantities known from real-time renormalization group methods for stationary quantities. We propose the study of a delay time as a characteristic time scale. We apply the method to the interacting resonant level model and calculate the nonlinear adiabatic charge response to time-dependent gate voltages, tunneling barriers and Coulomb interaction. The dot charge delay time is found to be given by a unique expression in all cases, in contrast to the capacitance and the charge relaxation resistance. We discuss the observability of the effects in molecular systems and cold-atom setups.

**Introduction.** – Adiabatic transport through quantum dots associated with a slow cyclic time dependence of the system parameters has generated a lot of interest in recent years, particularly in connection with quantum pumps [1–4] and mesoscopic capacitors [5, 6], see also Ref. [7]. For noninteracting systems the scattering formalism is a powerful tool to describe the adiabatic response [1, 2]. A central challenge in this field is the understanding of the influence of strong interactions as they typically occur in small quantum dots. Although general current formulas have been derived in terms of Green’s functions [8–10], their evaluation is quite difficult in the coherent regime at low temperature. Progress has been achieved in the perturbative regime of high temperature [11], where it was shown that pure interaction effects can be revealed by the adiabatic response which would be covered by more dominant effects in the steady state [11, 12]. These studies also included the properties of the  $RC$ -time in linear response [13]. In contrast, for interacting quantum dots at low temperature, a generic formalism providing the adiabatic time evolution in response to any parameter in linear or nonlinear response is not yet available. So far, quantum pumping has been studied for special models, like e.g. the 2-channel Kondo model in the strong coupling regime [9], the Kondo model at the exactly solvable Toulouse point [14], and the single-

impurity Anderson model within slave-boson mean-field approximation [15]. In addition, the research on interaction effects in mesoscopic capacitors has concentrated on the special case of linear charge response to an external AC gate voltage by using the standard relation to the equilibrium density-density correlation function. Here, the main object of interest was the charge relaxation resistance  $R$  defined by expanding the charge response  $\delta Q$  in the external frequency  $\omega$  via

$$\delta Q = (C + i\omega RC^2)\delta\mathcal{V} \quad , \quad (1)$$

where  $Q(t) = \delta Q e^{-i\omega t}$  denotes the charge,  $C$  is the static quantum capacitance and  $\mathcal{V}(t) = \delta\mathcal{V} e^{-i\omega t}$  defines the external AC gate voltage. For a single transport channel and provided that the Coulomb interaction is weak, a universal relaxation resistance  $R = h/2e^2$  was found in the coherent regime at zero temperature [5, 16]. For interacting metallic dots and the single-impurity Anderson model the Shiba relation was shown to be a powerful tool to analyze  $R$  and its universality [17–19]. Using bosonization, the influence of Luttinger-liquid leads on  $R$  has also been studied [20] and a numerical approach has been used away from the Fermi-liquid regime [21].

In this Letter we develop a general approach to deal with the adiabatic dissipative response, where the time scale of the external modulation  $\propto \omega^{-1}$  is much larger than the inverse of typical relaxation rates  $\Gamma_c$ , of a coherent quantum dot at low temperature including Coulomb interactions.

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We show that the adiabatic response can be calculated very efficiently by using quantum field theoretical methods in Liouville space developed in Refs. [22, 23] and generalized here for the case of time-dependent Hamiltonians (for approaches within Keldysh formalism see the review articles [7, 22] and the recent development [24]). We provide a general relationship of the adiabatic response to effective Liouvillians and vertices known from real-time renormalization group (RTRG) in the stationary limit with instantaneous time parametrization, based on powerful techniques for the calculation of Laplace-variable derivatives, recently used within the  $E$ -flow scheme of RTRG [25]. As a consequence, our formalism is suitable to any model which can be treated by RTRG, which is applicable for many generic models with charge and spin fluctuations [22]. Recent applications of RTRG cover the Kondo model for both weak [26–30] and strong [25] coupling, and the interacting resonant level model (IRLM) [31]. Most importantly, in contrast to previous research, our formalism allows for the adiabatic variation of *any* parameter in *non-linear* response, where no relation to equilibrium density-density correlation functions is possible and where certain identities like e.g. the Shiba relation are no longer applicable. Therefore, going beyond previous studies of the linear response to an external gate voltage, we also study the response to other parameters, like the tunneling coupling or the Coulomb interaction, which experimentally can be either realized intentionally, or indirectly induced by the gate voltage. We even cover the regime of non-linear response, motivated by recent works on mesoscopic capacitors in the nonlinear driving regime [32]. Instead of the linear response formula (1) for the charge variation by an external gate voltage, we decompose the dynamics of any observable  $A$ , with a nonvanishing instantaneous contribution and its adiabatic correction in response to any parameter, as  $A(t) = A^{(i)}(t) + A^{(a)}(t)$ . The central quantity of our interest is the delay time scale  $\tau_A$  for the expectation value  $A$ , defined by

$$\tau_A = |A^{(a)}/\dot{A}^{(i)}|, \quad (2)$$

which describes the delay of the full solution comparing to the instantaneous one. For  $A \equiv \hat{Q}$  in linear response to a time-dependent gate voltage, it is equivalent to the  $RC$ -time. In general  $\tau_A$  can be quite different from typical relaxation times, depending on the observable, the type of excitation and its amplitude, and it is of fundamental interest to understand its dependence on interactions.

We use our method to consider the IRLM with a single lead, which constitutes a minimal model for the mesoscopic capacitor with one single-particle level, where strong correlations are induced by a local Coulomb interaction between the dot and the lead. Recently, the IRLM has been extensively used to study nonequilibrium transport through interacting quantum dots [31, 33–36], including the dynamics of the time evolution into the stationary state [31]. We calculate the nonlinear adiabatic charge response and the delay time  $\tau_Q$ , including renormalization

effects of the tunneling enhanced by correlations. Importantly, we find that the functional form of the charge delay time  $\tau_Q$  is robust against the choice of the time-dependent parameter even in nonlinear response, whereas the capacitance  $C$  or the relaxation resistance  $R$  get a more complex form when the tunneling or the Coulomb interaction are varied. Finally, we analyze further possible experimental implementations of the predicted results for the IRLM with time-dependent parameters, namely via the Anderson-Holstein model in molecular electronics or via the spin-boson model in cold-atom setups.

**Method.** – We start from a general Hamiltonian  $H(t) = H_{\text{res}} + \sum_{\alpha} \mu_{\alpha}(t) \hat{N}_{\alpha} + H_{\text{dot}}(t) + V(t)$  of an interacting quantum dot coupled to noninteracting fermionic reservoirs with time-dependent chemical potentials  $\mu_{\alpha}(t)$  and a flat d.o.s. of width  $2D$  via the coupling  $V(t)$ . Generalizing the Liouvillian approach of Ref. [22] to the case of time-dependent Hamiltonians, one finds that the dissipative dynamics of the reduced density matrix  $\rho(t)$  of the dot can be described by the effective Liouvillian equation

$$i\partial_t \rho(t) = \int_{t_0}^t L(t, t') \rho(t') dt', \quad (3)$$

where  $L(t, t')$  is the effective dot Liouvillian obtained by integrating out the reservoirs ( $\hbar = 1$ ). At the initial time  $t_0$  the total density matrix factorizes into an arbitrary dot and an equilibrium reservoir part. Since we are only interested in the asymptotic dynamics we set  $t_0 = -\infty$  below. Following Ref. [22], the effective Liouvillian can be calculated diagrammatically, where each diagram of order  $O(V^n)$  consists of a product of vertices  $G(t_i)$ ,  $t_1 > \dots > t_n$ , with effective dot propagators  $\Pi(t_i, t_{i-1})$  in between. In addition, the information of the Fermi distribution and the d.o.s. of the reservoirs is contained in time-independent reservoir contractions between the vertices. Using the formal definition  $G(t, t') = G(t) \delta(t - t' - 0^+)$ , we find that each term can be written in terms of a generalized convolution in time space as  $(G \circ \Pi \circ \dots \circ \Pi \circ G)(t, t')$ , where  $(A \circ B)(t, t') \equiv \int_{t'}^t d\tau A(t, \tau) B(\tau, t')$ . Introducing the partial Laplace transform  $A(t; E) = \int_{-\infty}^t dt' e^{i(E+i0)(t-t')} A(t, t')$ , we get

$$(A_1 \circ A_2 \circ \dots \circ A_n)(t; E) = e^{\mathcal{D}} A_1(t; E) \dots A_n(t; E) = e^{i \sum_{j>k} \partial_{E_j} \partial_{t_k}} A_1(t_1, E_1) \dots A_n(t_n, E_n) \Big|_{E_j=E, t_k=t}. \quad (4)$$

The special differential operator  $\mathcal{D} = i\partial_E^{\text{left}} \partial_t^{\text{right}}$  prescribes the energy derivative to act left to the time derivative. This rule is a natural generalization to Laplace space of analog identities in Fourier space used for gradient expansions in the Keldysh formalism [37]. Formally, it allows for the straightforward application of the Liouvillian approach to time-dependent Hamiltonians, with the difference that the exponential differential operator has to be taken beforehand. In the adiabatic case, the exponential can be

expanded in  $\partial_E \partial_t \sim \frac{\omega}{\Gamma_c} \ll 1$ , leading to an expansion of the effective Liouvillian,

$$L(t; E) = L^{(i)}(t; E) + L^{(a)}(t; E) + \dots \quad (5)$$

Here,  $L^{(i)}(t; E)$  denotes the instantaneous part, where the time  $t$  enters only parametrically via the external parameters, and  $L^{(a)}(t; E)$  is the first adiabatic correction, which is linear in the time derivatives of the external parameters.

Once the effective Liouvillian  $L(t; E)$  is known up to the adiabatic correction, one can use it in eq. (3) which reads

$$i\partial_t \rho(t) = (L \circ \rho)(t; 0) = e^{i\partial_E^L \partial_t^R} L(t; E) \rho(t) \Big|_{E=0} \quad (6)$$

in the mixed  $(t; E)$ -representation. Expanding  $\rho(t) = \rho^{(i)}(t) + \rho^{(a)}(t) + \dots$  analogously to eq. (5), we find by comparing equal powers in the external frequency

$$L^{(i)} \rho^{(i)} = 0 \quad , \quad \text{Tr} \rho^{(i)} = 1 \quad , \quad \text{Tr} \rho^{(a)} = 0 \quad , \quad (7)$$

$$L^{(i)} \rho^{(a)} + L^{(a)} \rho^{(i)} - i(1 - \partial_E L^{(i)}) \partial_t \rho^{(i)} = 0 \quad . \quad (8)$$

In all arguments of  $L^{(i/a)}$  and  $\partial_E L^{(i)}$ ,  $E = 0$  has to be taken. From these equations the instantaneous density matrix  $\rho^{(i)}(t)$  and the first adiabatic correction  $\rho^{(a)}(t)$  can be determined. We emphasize that this approach is even applicable in nonlinear response in the amplitude of the external perturbations, i.e. only the time scale of the external modulation needs to be large enough. Furthermore, it allows for an adiabatic modulation of any parameter of the Hamiltonian and is not restricted to a time-dependent gate voltage.

The algebra of (7) and (8) can be easily evaluated for quantum dots with two accessible states. If additional conservation laws are present (as, *e.g.*, charge conservation in the IRLM or spin- $S_z$  conservation in the Kondo model), the nonvanishing matrix elements of the Liouvillian can be written as

$$L_{\bar{s}\bar{s},ss} = -L_{ss,ss} = i\Gamma_s = i\Gamma/2 + is\Gamma' \quad , \quad L_{s\bar{s},s\bar{s}} = \epsilon_s \quad , \quad (9)$$

where  $s \equiv \pm$  denotes the two states and  $\bar{s} = -s$ . At  $E = 0$  we get from (7) and (8) that the instantaneous density matrix is diagonal  $\rho_s^{(i)} = \Gamma_s^{(i)}/\Gamma^{(i)} = 1/2 + s\Gamma'^{(i)}/\Gamma^{(i)}$  and the adiabatic correction fulfills  $\rho_+^{(a)} = -\rho_-^{(a)}$  with

$$\rho_+^{(a)} = \frac{1}{\Gamma^{(i)}} \left\{ \Gamma'^{(a)} - \frac{\Gamma'^{(i)}}{\Gamma^{(i)}} \Gamma^{(a)} - (1 + i\partial_E \Gamma^{(i)}) \partial_t \left( \frac{\Gamma'^{(i)}}{\Gamma^{(i)}} \right) \right\} \quad (10)$$

Below we use this result to evaluate the adiabatic response for the IRLM.

**Calculation of  $L^{(a)}$ .** – We now turn to the central issue of how to relate the adiabatic correction  $L^{(a)}(t; E)$  to the instantaneous quantities known from RTRG. Using (4), we can formally write  $L^{(a)}(t; E) = \mathcal{D} L^{(i)}(t; E) = i\partial_E^{\text{left}} \partial_t^{\text{right}} L^{(i)}(t; E)$ . The representation of  $L^{(i)}(t; E)$  by its diagrammatic expansion specifies what “left” and

“right” means for the derivatives with respect to  $E$  and  $t$ . As a first step, we represent the derivative  $\partial_E L^{(i)}$  by effective vertices and propagators, using a method developed in Ref. [25]. For generic models with two types of vertices, namely single (*e.g.* tunneling) and double (*e.g.* Coulomb interaction, exchange, etc.), we decompose it into two contributions in leading order and find

$$\begin{aligned} \partial_E L^{(i)}(t; E) &= \partial_E L_{\Gamma}^{(i)}(t; E) + \partial_E L_U^{(i)}(t; E) \\ &= \text{[diagram: single vertex with red slash]} + \frac{1}{2} \text{[diagram: double vertex with red slash]} \quad . \quad (11) \end{aligned}$$

The diagrammatic rules are explained in detail in Refs. [22, 25]. The single (double) circles represent full effective single (double) vertices with effective propagators  $\Pi^{(i)}(t; E) = \frac{1}{E - L^{(i)}(t; E)}$  in between (the Laplace variable is shifted by the frequencies and chemical potentials of all reservoir contractions crossing over the propagator). The left slash indicates  $\partial_E$  and the grey (green, color online) line represents the reservoir contraction given by the antisymmetric part  $f(\omega) - \frac{1}{2}$  of the Fermi distribution function. All possible diagrams for  $\partial_E L^{(i)}$  can be classified by the number of lines over the propagator containing a derivative. In the next step we perform the time derivative  $i\partial_t$  right to the energy derivative. The energy derivative is then shifted by partial integration to the reservoir contraction (indicated by a (blue) cross) [25]. This yields

$$\begin{aligned} L_{\Gamma}^{(a)}(t; E) &= \text{[diagram: single vertex with red slash and blue cross]} + \text{[diagram: single vertex with red slash]} \\ &= - \text{[diagram: single vertex with red slash and blue cross]} + \text{[diagram: single vertex with red slash]} \quad , \quad (12) \end{aligned}$$

$$L_U^{(a)}(t; E) = \frac{1}{2} \text{[diagram: double vertex with red slash and blue cross]} + \frac{1}{2} \text{[diagram: double vertex with red slash]} \quad , \quad (13)$$

where the right slash represents  $i\partial_t$  and the hat indicates the differential operator  $\mathcal{D} = i\partial_E^{\text{left}} \partial_t^{\text{right}}$ . The frequency integral in both diagrams of (12) is well-defined in the wide-band limit, so (12) provides an explicit expression for the adiabatic correction containing the tunneling vertices in terms of renormalized vertices and propagators. In contrast, the frequency integrals in (13) are logarithmically divergent. We therefore take a second derivative with respect to  $E$ , yielding an RG equation for the adiabatic part,  $L_U^{(a)}(t; E)$ , after partial integration. This contains the double vertices

$$\partial_E L_U^{(a)}(t; E) = \frac{1}{2} \text{[diagram: double vertex with red slash and blue cross]} - \frac{1}{2} \text{[diagram: double vertex with red slash]} \quad . \quad (14)$$

Eqs. (12) and (14) are the final results for the evaluation of adiabatic corrections of the Liouvillian in leading order, based on the instantaneous values of the renormalized vertices and Liouvillian, which are obtained from RTRG<sup>1</sup>. For the adiabatic part of the propagator, appearing in the second diagram of (12) and (14) each, we insert

<sup>1</sup>Provided that the frequency integrals converge, we note that our results can even be applied to a frequency-dependent d.o.s. in the leads. Otherwise, another derivative with respect to  $E$  may be required.

$\Pi^{(a)} = \Pi^{(i)} L^{(a)} \Pi^{(i)} + (\partial_E \Pi^{(i)})(i\partial_t L^{(i)}) \Pi^{(i)}$ . The first term does however not contribute to the adiabatic propagator in leading order.

An interesting question is whether derivatives with respect to the Laplace and time variable commute in leading order, i.e. whether the adiabatic correction to the effective Liouvillian, eqs. (12) and (14), can be written as

$$L_{\Gamma/U}^{(a)}(t; E) \stackrel{?}{=} \frac{1}{2} i\partial_E \partial_t L_{\Gamma/U}^{(i)}(t; E). \quad (15)$$

A similar relation was investigated so far only for noninteracting systems [38]. To analyze its validity we introduce the complementing differential operator  $\mathcal{D}' = i\partial_E^{\text{right}} \partial_t^{\text{left}}$ , where the energy derivative is taken *right* to the time derivative. Analogously to (12) and (14) one finds in leading order

$$\mathcal{D}' L_{\Gamma}^{(i)}(t; E) = - \text{diagram} + \text{diagram}, \quad (16)$$

$$\partial_E \mathcal{D}' L_U^{(i)}(t; E) = \frac{1}{2} \text{diagram} - \frac{1}{2} \text{diagram}. \quad (17)$$

The inverted hat represents the differential operator  $\mathcal{D}'$ . Using  $i\partial_E \partial_t = \mathcal{D} + \mathcal{D}'$ , we can write  $\mathcal{D} = \frac{1}{2} i\partial_E \partial_t + \frac{1}{2}(\mathcal{D} - \mathcal{D}')$  and, thus, the correction to eq. (15) for  $L_{\Gamma}^{(a)}$  ( $\partial_E L_U^{(a)}$ ) is given by half the difference of (12) and (16) ((14) and (17)). We first address the second diagrams on the r.h.s. of these equations: their differences involve the expression

$$\begin{aligned} \frac{1}{2}(\mathcal{D} - \mathcal{D}') \Pi^{(i)} &= \Pi^{(i)} \left( \frac{1}{2}(\mathcal{D} - \mathcal{D}') L^{(i)} \right) \Pi^{(i)} \\ &+ \frac{1}{2} \left\{ (\partial_E \Pi^{(i)})(i\partial_t L^{(i)}) \Pi^{(i)} - \Pi^{(i)}(i\partial_t L^{(i)})(\partial_E \Pi^{(i)}) \right\} \end{aligned} \quad (18)$$

for the propagator. Here, the first term on the r.h.s. can be neglected in leading order, whereas the second one is only zero if the Liouvillian and its time and energy derivative commute. For special cases this is indeed possible: it follows trivially for blocks where the Liouvillian is diagonal, as e.g. the  $2 \times 2$ -block  $L_{s\bar{s}, s'\bar{s}'}^{(i)} = \delta_{ss'} \epsilon_s^{(i)}$  of eq. (9). For 2-level systems with conservation laws, see eq. (9), it holds also for the block  $L_{ss, s's'}^{(i)}$  since the zero eigenvalue of the Liouvillian can be omitted in a propagator standing left to a vertex averaged over the Keldysh indices [22]. Therefore, for this block one can replace the Liouvillian by its nonzero eigenvalue  $-i\Gamma^{(i)}(t; E)$  and the second term on the r.h.s. of (18) is again zero. If this is given (or if the term can be neglected in leading order for certain models), we can write the correction to eq. (15) generically as

$$\begin{aligned} L_{\Gamma}^{(a)}(t; E) &= \frac{1}{2} i\partial_E \partial_t L_{\Gamma}^{(i)}(t; E) \\ &+ \frac{1}{2} \text{diagram} - \frac{1}{2} \text{diagram}, \quad (19) \end{aligned}$$

$$\begin{aligned} \partial_E L_U^{(a)}(t; E) &= \partial_E \left\{ \frac{1}{2} i\partial_E \partial_t L_U^{(i)}(t; E) \right\} \\ &+ \frac{1}{4} \text{diagram} - \frac{1}{4} \text{diagram}. \quad (20) \end{aligned}$$

From this result we observe another condition for the validity of (15), namely that it should not matter whether the right or the left vertex is differentiated with respect to time, i.e. the two vertices should be equivalent. Whether this is the case, depends on the algebra of the model under consideration. For noninteracting systems one can take bare vertices and the reservoir indices of the two vertices have to be the same due to the reservoir contractions connecting them. In this case the condition is fulfilled if the vertices do not depend on the level index of the dot states, e.g. through differently time-dependent coupling to different leads [39]. For interacting systems, the validity of (15) is more restrictive. The renormalized vertices can be quite different from the bare ones and the vertices get an additional dependence on the Laplace variable  $E$  which is shifted by the chemical potentials of the reservoir lines crossing over the propagator standing left to that vertex. As a consequence, the two vertices are never equivalent in the presence of a bias voltage and correction terms definitely occur for time-dependent voltages. As discussed below, for the particular case of the IRLM with one single reservoir, correction terms to eq. (15) do not appear in leading order.

**Results.** — We use the above developed method to analyze the response of a mesoscopic capacitor at zero temperature, described by the IRLM, where  $H_{\text{res}} = \sum_k \epsilon_k a_k^\dagger a_k$  describes a noninteracting reservoir with flat d.o.s.  $\nu$  of band width  $2D$ ,  $H_{\text{dot}}(t) = \epsilon(t)c^\dagger c$  denotes a spinless single-level quantum dot with time-dependent level position  $\epsilon(t)$ , and

$$\begin{aligned} V(t) &= \sqrt{\frac{\Gamma_0(t)}{2\pi\nu}} \sum_k (c^\dagger a_k + h.c.) \\ &+ \frac{U(t)}{\nu} \sum_{kk'} (c^\dagger c - 1/2) a_k^\dagger a_{k'} \end{aligned} \quad (21)$$

is the dot-reservoir coupling with the bare time-dependent tunneling rate  $\Gamma_0(t)$  and the time-dependent dimensionless Coulomb interaction  $U(t)$ .

As shown above, we can evaluate the adiabatic response from eq. (10), where  $\Gamma^{(a)}$  and  $\Gamma'^{(a)}$  can be extracted from eqs. (19) and (20) together with the RTRG results for the instantaneous vertices and the Liouvillian derived in Ref. [31]. For  $E = 0$  and leading order in  $U$ , the results of Ref. [31] read

$$\Gamma = \Gamma_0 \left( \frac{D}{|\epsilon - i\Gamma/2|} \right)^{2U}, \quad \partial_E \Gamma = i \frac{U\Gamma^2}{\epsilon^2 + (\frac{\Gamma}{2})^2}, \quad (22)$$

$$\Gamma' = -\frac{\Gamma}{\pi} \arctan \frac{\epsilon}{\Gamma/2}, \quad \partial_E \Gamma' = -\frac{i}{\pi} \frac{\Gamma\epsilon}{\epsilon^2 + (\frac{\Gamma}{2})^2}, \quad (23)$$

where we have omitted the index  $(i)$  for the instantaneous quantities. Furthermore, the analysis in Ref. [31] shows that the Coulomb vertex is zero in leading order for the Liouvillian elements containing  $\Gamma$  and  $\Gamma'$ . Therefore eq. (19)



is sufficient to evaluate  $\Gamma^{(a)}$  and  $\Gamma'^{(a)}$ . Inserting the algebra for the instantaneous tunneling vertices into eq. (19), one finds that eq. (15) is valid for the calculation of  $\Gamma'^{(a)}$ , whereas for  $\Gamma^{(a)}$  a correction term occurs proportional to  $\partial_t U$ . This yields the total result

$$\Gamma^{(a)} = -\frac{U}{2}\partial_t \frac{\Gamma^2}{\epsilon^2 + (\frac{\Gamma}{2})^2}, \quad \Gamma'^{(a)} = \frac{1}{2\pi}\partial_t \frac{\Gamma\epsilon}{\epsilon^2 + (\frac{\Gamma}{2})^2}. \quad (24)$$

Since  $\partial_E \Gamma, \Gamma^{(a)} \sim O(U)$  we can neglect them in leading order in eq. (10) and, by inserting (22) to (24) into (10), we find after a straightforward analysis for the charge response given by  $Q = e\rho_+$ ,

$$\dot{Q}^{(i)} = C_0 \Gamma \partial_t \frac{\epsilon}{e\Gamma}, \quad Q^{(a)} = -R_0 C_0^2 \Gamma \partial_t \frac{\epsilon}{e\Gamma}, \quad (25)$$

where  $R_0 = \frac{\hbar}{2e^2}$  and  $C_0 = \frac{e^2}{2\pi} \frac{\Gamma}{\epsilon^2 + (\Gamma/2)^2}$ . In the special case of linear response and when only  $\epsilon$  is varied with time,  $C = C_0$  is the static capacitance and  $R = R_0$  the universal relaxation resistance, in agreement with (1). In contrast, when  $\Gamma$  is varied with intent or via an accidental (but experimentally unavoidable) gate voltage dependence of  $\Gamma_0$  or  $U$ , we obtain in linear response eq. (1) with

$$C = C_0 \left(1 - \frac{\epsilon}{\Gamma} \frac{\partial \Gamma}{\partial \epsilon}\right), \quad R = \frac{R_0 C_0}{C}, \quad (26)$$

where  $\frac{\partial \Gamma}{\partial \epsilon} \approx \frac{\Gamma}{\Gamma_0} \frac{\partial \Gamma_0}{\partial \epsilon} + 2\Gamma \frac{\partial U}{\partial \epsilon} \log \frac{D}{|\epsilon - i\Gamma/2|}$ . As a consequence,  $C$  and  $R$  are very sensitive to the variation of other parameters, and logarithmic terms due to renormalization effects occur, if the Coulomb interaction  $U$  varies with time.

In this general case, where also the renormalized  $\Gamma$  varies with time, we propose to analyze the time scale  $\tau_Q$ . From (2) and (25) we get

$$\tau_Q = \left| \frac{Q^{(a)}}{\dot{Q}^{(i)}} \right| = \frac{\Gamma/2}{\epsilon^2 + (\Gamma/2)^2} = R_0 C_0, \quad (27)$$

which is of  $O(\Gamma^{-1})$  close to resonance  $\epsilon \sim \Gamma$  and of  $O(\Gamma/\epsilon^2)$  away from resonance. This result holds for *any* variation of  $\epsilon$ ,  $\Gamma_0$  and  $U$  and is also valid in nonlinear response. Interaction effects enter only weakly via the renormalized  $\Gamma$  given by (22). Importantly,  $\tau_Q$  reveals the static capacitance  $C_0$  for a pure change of the gate voltage in linear response, with the advantage that  $\tau_Q$  can be determined in the presence of the variation of any parameter.

The experimentally accessible time scale  $\tau_Q$  is thus an interesting quantity, which, for the case of the IRLM, is stable for the variation of any parameter in linear or nonlinear response. We note that this time scale can vary quite drastically if other observables or other models are studied. E.g., the time scales  $\tau_Q$  and  $\tau_I$ , when  $Q$  is replaced by the current  $I = \dot{Q}$ , are in general the same only in linear response. For the IRLM, the time scale  $\tau_I$  shows similar logarithmic renormalizations in nonlinear response as they occur in  $C$  and  $R$  for time varying  $U$ .

**Realizations.** – Several experimental realization of the IRLM exist, where the different parameters can be modulated in a controlled way. As we outline here, the applicability of the IRLM is not limited to the description of an interacting quantum dot, but allows the observation of the predicted effects for various physical systems. First, we show that the low-energy physics of the IRLM is equivalent to the one of the Anderson-Holstein model, as first predicted in Ref. [40]. This model is widely used in molecular electronics [41] and describes a single-level molecular quantum dot, having a vibrational degree of freedom with frequency  $\Omega$  coupled linearly to the charge of the dot

$$H_{\text{dot}} = \epsilon_M c^\dagger c + \Omega b^\dagger b - \lambda \Omega (b + b^\dagger) c^\dagger c, \quad (28)$$

$$V = \sqrt{\frac{\Gamma_M}{2\pi\nu}} \sum_k (c^\dagger a_k + h.c.). \quad (29)$$

The parameters  $\epsilon_M$ ,  $\Gamma_M$ ,  $\lambda$  and  $\Omega$  can be related to the effective parameters  $\epsilon$ ,  $\Gamma_0$  and  $U$  of the IRLM. Applying a Lang-Firsov transformation [42], the coupling to the vibrational mode can be incorporated into the tunneling, leading to an effective level position,  $\epsilon = \epsilon_M - \lambda^2 \Omega$ , and tunneling rate,  $\Gamma_0 = \Gamma_M e^{-\lambda^2}$  [42, 43]. If the vibration frequency  $\Omega$  is large compared to the other energy scales, the virtual intermediate states between the tunneling sequences with one or more bosons can be integrated out. This produces terms with  $n \geq 2$  lead operators in the effective Hamiltonian. At large  $\lambda$ , all cotunneling processes with  $n > 2$  are exponentially suppressed, while the two-particle processes enter as an effective interaction. Hence, by integrating out all vibrational modes the Anderson-Holstein model can be mapped onto the IRLM with effective interaction,  $U = \frac{\Gamma_M}{2\pi\lambda^2\Omega}$ , with  $\Omega \sim D$ . This is in agreement with Ref. [40], where it was shown numerically that this formula has even a broader range of applicability. A modulation of the tunneling barriers is always accompanied by a modulation of the effective interaction  $U$ , since it is proportional to the tunneling rate  $\Gamma_M$ . In this case, our results predict that logarithmic corrections appear for  $C$  and  $R$ , whereas the time scale  $\tau_Q$  only depends on  $\epsilon$  and  $\Gamma$  via eqs. (27) and (22). The Holstein coupling in the Hamiltonian allows for the observation of the dot charge via the displacement of the dot  $\sim \langle b + b^\dagger \rangle$ .

Finally, our results can be used to extract information on the relaxation behavior of systems described by the spin-boson model, namely, two-level dissipative systems connected to a large ensemble of oscillators

$$H = \frac{\epsilon}{2} \sigma_z - \frac{\Delta}{2} \sigma_x + \sum_q \omega_q b_q^\dagger b_q + \frac{\sigma_z}{2} \sum_q g_q (b_q + b_q^\dagger). \quad (30)$$

The spin-boson model can be implemented by a Bose condensate of atoms trapped by a focused laser beam [44]. Such ultracold gases in optical lattices provide experimental realizations for theoretical models with remarkably independent tunability of parameters including the interaction strength, in contrast to usual semiconductor quantum dot setups. The system's behavior depends crucially

on the spectral coupling function. For the ohmic case, i.e. when the coupling constant obeys  $\sum_q g_q^2 \delta(\omega - \omega_q) = 2\alpha\omega e^{-\omega/D}$ , the spin-boson model can be mapped onto the IRLM if  $\alpha \approx 1/2$  (close to the Toulouse limit) [45, 46]. The effective IRLM parameters are  $U = 1 - \sqrt{2\alpha}$  and  $\Gamma_0 = \Delta^2/D$ . Changing the coupling of the Bose condensate to the spin via  $\alpha$  one generates a time-dependent effective interaction  $U$ . The resulting response  $\langle S_z \rangle$  of the spin, identified with  $(\rho_+ - \rho_-)/2$  in the effective IRLM, allows for the determination of the interesting time scale  $\tau_{S_z}$ , given by (27). Especially in the biased case, where  $\tau_{S_z} \sim \Gamma/\epsilon^2$ , this time scale is expected to differ significantly from typical relaxation rates  $\Gamma$  and  $\Gamma/2$  for the diagonal and nondiagonal components of the density matrix [31, 47].

**Conclusions.** — In this Letter we provide a generic relation of the adiabatic response to real-time RG results for the stationary case. The presented scheme allows for the variation of any parameter in linear or nonlinear response and provides criteria when the adiabatic correction to the Liouvillian can be calculated directly via energy and time derivatives of the instantaneous one. We suggest a delay time as an interesting time scale and show for the IRLM that its expression is robust against the choice of time-dependent parameters and their amplitude. We confirm the universality of the AC relaxation resistance, unless a time dependence of tunneling and interaction is present, revealing logarithmic renormalizations due to charge fluctuations. We proposed different setups in molecular electronics and cold-atom systems to observe the effects experimentally.

\* \* \*

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## REFERENCES

- [1] BÜTTIKER M., THOMAS H., and PRÊTRE A., *Z. Phys. B*, **94** (1994) 133.
- [2] BROUWER P. W., *Phys. Rev. B*, **58** (1998) R10135.
- [3] POTHIER H. *et al.*, *Europhys. Lett.*, **17** (1992) 249.
- [4] GIAZZOTTO F. *et al.*, *Nat. Phys.*, **7** (2011) 857.
- [5] BÜTTIKER M., THOMAS H., and PRÊTRE A., *Phys. Lett. A*, **180** (1993) 364.
- [6] GABELLI J. *et al.*, *Science*, **313** (2006) 499.
- [7] ANDERGASSEN S. *et al.*, *Nanotechnology*, **21** (2010) 272001.
- [8] SPLETTSTOESSER J. *et al.*, *Phys. Rev. Lett.*, **95** (2005) 246803.
- [9] SELA E. and OREG Y., *Phys. Rev. Lett.*, **96** (2006) 166802.
- [10] FIORETTO D. and SILVA A., *Phys. Rev. Lett.*, **100** (2008) 236803.
- [11] SPLETTSTOESSER J. *et al.*, *Phys. Rev. B*, **74** (2006) 085305.
- [12] RECKERMANN F., SPLETTSTOESSER J. and WEGEWIJS M. R., *Phys. Rev. Lett.*, **104** (2010) 226803.
- [13] SPLETTSTOESSER J. *et al.*, *Phys. Rev. B*, **81** (2010) 165318.
- [14] SCHILLER A. and SILVA A., *Phys. Rev. B*, **77** (2008) 045330.
- [15] AONO T., *Phys. Rev. Lett.*, **93** (2004) 116601.
- [16] NIGG S. E., LÓPEZ R. and BÜTTIKER M., *Phys. Rev. Lett.*, **97** (2006) 206804.
- [17] RODIONOV Y. I., BURMISTROV I. S. and IOSELEVICH A. S., *Phys. Rev. B*, **80** (2009) 035332.
- [18] MORA C. and LE HUR K., *Nat. Phys.*, **6** (2010) 697.
- [19] FILIPPONE M., LE HUR K. and MORA C., *Phys. Rev. Lett.*, **107** (2011) 176601.
- [20] HAMAMOTO Y. *et al.*, *Phys. Rev. B*, **81** (2010) 153305.
- [21] LEE M. *et al.*, *Phys. Rev. B*, **83** (2011) 201304.
- [22] SCHOELLER H., *Eur. Phys. J. Special Topics*, **168** (2009) 179.
- [23] LEIJNSE M. and WEGEWIJS M. R., *Phys. Rev. B*, **78** (2008) 235424.
- [24] D. Kennes and V. Meden, *arXiv:1204.2100*.
- [25] PLETYUKHOV M. and SCHOELLER H., *arXiv:1201.6295*.
- [26] SCHOELLER H. and REININGHAUS F., *Phys. Rev. B*, **80** (2009) 045117.
- [27] PLETYUKHOV M., SCHURICHT D. and SCHOELLER H., *Phys. Rev. Lett.*, **104** (2010) 106801.
- [28] SCHURICHT D. and SCHOELLER H., *Phys. Rev. B*, **80** (2009) 075120.
- [29] PLETYUKHOV M. and SCHURICHT D., *Phys. Rev. B*, **84** (2011) 041309.
- [30] HÖRIG C. B. M., SCHURICHT D. and ANDERGASSEN S., *Phys. Rev. B*, **85** (2012) 054418.
- [31] KARRASCH C. *et al.*, *Europhys. Lett.*, **90** (2010) 30003; ANDERGASSEN S. *et al.*, *Phys. Rev. B*, **83** (2011) 205103.
- [32] FEVE G. *et al.*, *Science*, **316** (2007) 1169; MOSKALETS M., SAMUELSSON P. and BÜTTIKER M., *Phys. Rev. Lett.*, **100** (2008) 08660.
- [33] MEHTA P. and ANDREI N., *Phys. Rev. Lett.*, **96** (2006) 216802; *cond-mat/0703426* (Erratum).
- [34] BOULAT E. and SALEUR H., *Phys. Rev. B*, **77** (2008) 033409.
- [35] BOULAT E. SALEUR H. and SCHMITTECKERT P., *Phys. Rev. Lett.*, **101** (2008) 140601.
- [36] KARRASCH C. *et al.*, *Phys. Rev. B*, **81** (2010) 125122.
- [37] RAMMER J. and SMITH H., *Rev. Mod. Phys.*, **58** (1986) 323.
- [38] MOSKALETS M. and BÜTTIKER M., *Phys. Rev. B*, **69** (2004) 205316.
- [39] MOSKALETS M. and BÜTTIKER M., *Phys. Rev. B*, **78** (2008) 035301.
- [40] EIDELSTEIN E., GOBERMAN D. and SCHILLER A., in preparation.
- [41] BOESE D. and SCHOELLER H., *Europhys. Lett.*, **54** (2001) 668; WEGEWIJS M. R. and NOWACK K. C., *New J. of Physics*, **7** (2005) 239.
- [42] LANG I. G. and FIRSOV YU. A., *Sov. Phys. JETP*, **16** (1963) 1301.
- [43] KOCH T. *et al.*, *J. Phys.: Conf. Ser.*, **220** (2010) 012014.
- [44] RECATI A. *et al.*, *Phys. Rev. Lett.*, **94** (2005) 040404.
- [45] TSVELICK A. M. and WIEGMANN P. B., *Adv. in Physics*,

**32** (1983) 453.

[46] LEGGETT A. J. *et al.*, *Rev. Mod. Phys.*, **59** (1987) 1.

[47] EGGER R., GRABERT H. and WEISS U., *Phys. Rev. E*,  
**55** (1997) R3809.